REPORT DOCUMENTATION PAGE

Form Approved OMB No 0704-0188

1. AGENCY USE ONLY (Leave blank)

2. REPORT DATE: 1990

3. REPORT TYPE AND DATES COVERED: Final 1 Oct 84 - 30 Sep 89

4. TITLE AND SUBTITLE: Percolation & Low Density Materials: Theory & Applications

5. FUNDING NUMBERS: DAAG29-84-K-0136

6. AUTHOR(S): William Klein

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)

Boston University
Department of Physics
Boston, Massachusetts 02215

8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)

U. S. Army Research Office
P. O. Box 12211
Research Triangle Park, NC 27709-2211

10. SPONSORING/MONITORING AGENCY REPORT NUMBER: ARO 21654.9-PH

11. SUPPLEMENTARY NOTES

The view, opinions and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy, or decision, unless so designated by other documentation.

12a. DISTRIBUTION/AVAILABILITY STATEMENT

Approved for public release; distribution unlimited.

12b. DISTRIBUTION CODE

13. ABSTRACT (Maximum 200 words)

In the past several years it has become increasingly clear that connectivity or percolation concepts play an important role in understanding the properties of these low-density materials. The full exploitation of this important link between material properties and connectivity has, until recently, been hampered by the scarcity of reliable methods to handle the complex connectivity problem and the lack of tractable models that retain the essential physics.

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14. SUBJECT TERMS: Nucleation, Complex Connectivity Problem, Spinodal Decomposition, Percolation Concepts, Low-Density Materials, Material Properties, Disordered Alloys, Diffusion, Elasticity.

15. NUMBER OF PAGES: 44

16. PRICE CODE

17. SECURITY CLASSIFICATION OF REPORT	18. SECURITY CLASSIFICATION OF THIS PAGE	19. SECURITY CLASSIFICATION OF ABSTRACT	20. LIMITATION OF ABSTRACT
UNCLASSIFIED	UNCLASSIFIED	UNCLASSIFIED	UL

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ARO 21654.9-PH

FINAL REPORT

to the

ARMY RESEARCH OFFICE

ARO Contract DAAG-29-84-K-0136

Submitted by:

Boston University
Center for Polymer Studies
Department of Physics
590 Commonwealth Avenue
Boston, Massachusetts 02215
Tel: 617/353-2617 Fax: 617/353-3783

Abstract

In the past several years it has become increasingly clear that connectivity or percolation concepts play an important role in understanding the properties of these low-density materials. The full exploitation of this important link between material properties and connectivity has, until recently, been hampered by the scarcity of reliable methods to handle the complex connectivity problem and the lack of tractable models that retain the essential physics. Substantial progress has been made and we have seen the beginning of important applications of percolation to several materials problems such as elasticity, nucleation, spinodal decomposition and diffusion in disordered alloys.

Contents

INTRODUCTION

During the period of this grant our work has focused on three broad problems. The first is the development of a cell position-space renormalization group approach for percolation that has as its primary advantages conceptual simplicity and systematic improvability. The basic idea of the method is to rescale occupation probabilities directly without any appeal to renormalizing an underlying statistical mechanical model Hamiltonian. This rescaling is carried out within a cell framework in which the sources of error inherent in this approximation scheme are clearly identifiable as the surface-to-volume ratio of the cell. We were thereby led to consider the large-cell limit, and for this purpose we developed a Monte Carlo renormalization procedure. Due to the sharpness of the underlying probability distributions, extreme accuracy can be obtained when only 10^3 out of the possible $2^{250,000}$ states of a 500×500 cell are sampled. Finally, in conjunction with finite-size scaling, the sequence of results of finite-cell renormalizations can be extrapolated to the infinite-cell limit. This method provides numerical accuracy that compares favorably with traditional techniques. The paper describing this work appeared in the list of the 100 most frequently cited physics papers of 1982. A second focus of our work has been the development of more general percolation models. Some are motivated by attempts to explain experimental data on diverse systems, such as dilute ferromagnets and gels. We have also considered models which have inherent theoretical interest, and which also provide further insights into the geometric structure of random systems. For example, we have studied various correlated percolation models, anisotropic and directed percolation, and percolation in a continuum. Our very recent work in directed percolation has shown that the interplay of concentration and orientational degrees of freedom in a random system leads to new types of critical behavior. This may have important ramifications for transport in random systems, such as electron hopping conductivity in strong fields, where directionality constraints are important. A third direction of our work involves the development of a simple geometric description for the structure of the "incipient" infinite cluster which forms at the percolation threshold. This structure directly controls the properties of any physical problem embedded on the infinite cluster; for example the flow of electrical current or the spread of magnetic correlations. The primary difficulty in visualizing a geometric picture of the percolating cluster is that no characteristic length scale exists–the cluster is self-similar on all length scales. Nevertheless we have developed a very simple theory based on identifying geometric quantities that unambiguously characterize the structure of the incipient infinite cluster. Our theory successfully describes recent experiments on dilute ferromagnets and provides further predictions amenable to experimental test.

A. Fractal Structure of Low Density Materials

Percolation is a model suitable to describe low density materials. Many properties of such materials strongly depend on the structure of the incipient infinite network. For example, the elastic modulus of polymer gels and vulcanized rubber, the electric resistance of a random resistor network, the diffusion of a fluid in porous media, communication in a network of connected stations, and order propagation in dilute ferromagnets all depend strongly on the structure of the infinite cluster.

How can one characterize the structure of the incipient infinite cluster? Many attempts have been made by introducing suitable models such as the links and nodes model (Skal and Shklovskii 1975, de Gennes 1976) and more recently the Sierpinski gasket (Gefen et al 1981). Although the models are rather simple and amenable to direct calculation, we now realize that they fail to describe the right behavior (Stanley and Coniglio 1983).

Just below the percolation threshold p_c, consider the typical cluster of linear dimension $\xi \sim (p - p_c)^{-\nu_p}$ where ν_p is the *connectedness length exponent*. If we imagine that each bond carries an unit electrical resistance and we apply a voltage between the two extreme points i and j separated by a distance of order ξ, the bonds will fall in two categories (Stanley 1977): (i) dead ends that do not carry the current (yellow bonds) and (ii) backbone bonds that do carry current. The backbone bonds are responsible for long range connectivity. For example, they support a shear stress in a polymer gel or rubber. The dead ends could be removed, thereby lowering the density of the material without changing the macroscopic properties such as the elasticity or the electrical conductivity. What is the structure of this fundamentally important backbone? Again there are two categories of bonds. The links (red bonds) are such that if one is cut the backbone breaks into two disconnected clusters. All other bonds (blue bonds) form blobs. We have made a systematic study of the statistics of these bonds based on exact results, series expansion and Monte Carlo simulations (Stanley and Coniglio 1983, Pike and Stanley 1981, Coniglio 1981,1982, Hong and Stanley 1983a). This analysis shows that all these bonds are critical

$$\begin{aligned} L &= \text{number of red bonds} \sim \xi^{1/\nu_p} \\ L_B &= \text{number of blue bonds} \sim \xi^{d_f} \qquad (1) \\ L_Y &= \text{number of yellow bonds} \sim \xi^{d_f}. \end{aligned}$$

Here d_f is the fractal dimensionality of the full incipient infinite cluster and $\bar{d}_f$ is the fractal dimensionality of the backbone. Note that $d_f = y_h$, the critical exponents associated with the "ghost field." If the backbone bonds were only made of cutting bonds as in the nodes and links model, we would have $L_B = L$ and consequently $\bar{d}_f/\nu_p = 1$. Analytical results show that this is true only for dimensionality $d = 1$ and 6. For intermediate values of d, $d_B/\nu_p > 1$. The more d_B/ν_p is larger than 1 the more relevant are the "blobs." Numerical calculations show that d_B/ν_p assumes its maximum values for $d = 2$. Based on the above analysis, we have suggested that above p_c the infinite cluster is made of nodes connected by a quasi one-dimensional chain made of links and blobs. The blobs themselves are made of chains made of links and blobs in a self similar way. We note that the number of cutting bonds diverge with a superuniversal exponent 1 when p_c is approached from below $L \sim (p_c - p)^{-1}$. This is true in any dimension and for any lattice.

Critical Phenomena in Random Systems

Dilute ferromagnets have received much attention recently both experimentally and theoretically (Birgeneau et al 1976,1980, Cowley et al 1980, Stauffer 1975, Stanley et al 1976, Lubensky 1977, Stephen and Grest 1977) because their comprehension is important for the general understanding of many other disordered systems. Consider, for example, an Ising model in which ferromagnetic bonds are randomly distributed with concentration p. If all the bonds are present ($p = 1$), the pure Ising model is recovered. As p decreases, the average ferromagnetic interaction also decreases. As a consequence, the critical temperature decreases and approaches zero at the percolation threshold p_c. Below this value, only finite clusters of ferromagnetic bonds are present, and therefore no ferromagnetic order is possible. The special point $Q(p = p_c, T = 0)$ is very intriguing as both connectivity and thermal fluctuation become critical. As this point is approached along the path $p \to p_c, T = 0$ no thermal fluctuations are present and the critical behavior is characterized by percolation exponents.

Much more interesting is the case when Q is approached along the path $T \to 0, p = p_c$. The experiments show two different behaviors according to the symmetry of the Hamiltonian that describes the system under investigation. For 2- and 3-dimensional Ising systems (discrete symmetry) $\nu_t = \nu_p$ giving therefore a crossover exponent $\phi = 1$. While for a 2-dimensional Heisenberg system (continuous symmetry) the most recent experimental data give $\phi = \nu_T/\nu_p = 1.48 \pm 0.15$. These data have been without a satisfactory explanation for a long time. We have studied at p_c the thermal critical behavior of two classes of models:

the q-state Potts model, which contains the Ising model as a particular case ($q = 2$), and the n-vector model, which contains as a particular case the Heisenberg model for $n = 3$.

Discrete symmetry (Potts model). By applying an exact renormalization group procedure near $T = 0$ (Coniglio 1981), we have found that the thermal correlation length exponent along direction $p = p_c, T \to 0$ is given by

$$\nu_T = \frac{\ln \xi}{\ln L}, \tag{2}$$

where $\xi \sim |p - p_c|^{-\nu_p}$ is the connectedness length and L is the number of red bonds in the incipient infinite cluster. Using the result found previously, i.e., $L \sim |p - p_c|^{-1}$ it follows that $\nu_T = \nu_p$ in agreement with the experimental data and ϵ-expansion (Stephen and Grest 1977, Wallace and Young 1978). The quantity L plays the role of an effective 1-dimensional length along which thermal information is transmitted. In fact the same renormalization group applied to a 1-dimensional length of L steps gives the same eigenvalue at $T = 0$ and therefore the same critical behavior near $T = 0$. Physically only the singly connected bonds contribute to L, because the spins in the blobs of multiply-connected bonds are strongly correlated at low temperatures, and therefore do not offer any "resistance" to the spread of thermal correlations.

Continuous symmetry (n-vector model with $n > 1$). Application of the same exact renormalization procedure near $T = 0$ shows that the effective 1-dimensional length along which thermal information is transmitted is given by the 1-dimensional resistance associated to the backbone L_R. Note that now the blobs do contribute to this effective length. The physical reason for this is that the spins in the blobs are not as strongly correlated as in the q-state Potts model, because of the low energy excitations of the spin waves. As a consequence, we find that $\nu_T = \nu_p / z_R$ where z_R is the critical exponent associated with the divergence of the resistance. Using the numerical data for z_R we find good agreement with the experimental data. In this way not only have we found numerical agreement with the experimental data, but we also have related these numerical quantities to the geometrical properties of the incipient infinite cluster.

B. Anisotropic Aspects of Low-Density Materials

Very recently, it has been recognized that anisotropy, or directionality constraints, can play a fundamental role in influencing the properties of random media. Examples include fluid flow in randomly porous media at high flow rates, strong-field hopping conduction in doped semiconductors, formation of a gel in a flowing solvent, and composite materials consisting of anisotropic elements such as fibers or oriented polymers. As an initial step in understanding the effect of directionality on random systems, we are carrying out a research program on a variety of models in which anisotropy effects play a fundamental role. One important example is ***directed*** percolation, a model in which directed bonds, or diodes, of a specified orientation randomly occupy a lattice (Kertész and Vicsek 1980, Obukhov 1980, Cardy and Sugar 1980, Dhar and Barma 1981, Kinzel and Yeomans 1981; for reviews, see e.g., Kinzel 1983, Redner 1983). This simple model appears to capture many of the intriguing physical phenomena found in the systems mentioned above. For example, directed percolation may be used to describe the non-linear conductivity and the phenomenon of ***negative*** resistance found in strong field hopping conduction processes (Böttger and Bryksin 1979, 1980, Van Lien and Shklovskii 1981).

Our work on directed percolation has also led us to introduce and study a more general model which incorporates both non-directed bonds (resistors), and directed bonds (diodes) of ***arbitrary*** orientation (Redner 1981, 1982a,b). The additional orientational degrees of freedom of the diodes gives rise to richer network behavior than that found in conventional percolation or in directed percolation. It should be stressed that the directionality constraint of the diodes drastically alters the basic physical features of the percolation models. Our recent work in this area represents some important first steps in understanding the ramifications of these directionality constraints.

A mean-field theory for directed percolation was constructed in order to understand the fundamental anisotropic nature of the transition (Redner 1982). The basic new idea is that fluctuations in a Landau-Ginzburg expansion of the free energy are accounted for by including both even and odd powers of the gradient. This stems from the fact that a symmetry-breaking occurs because of the preferred direction defined by the bias of the diodes. From this free energy, we find that correlations in the direction parallel to the orientation of the diodes are much longer-ranged than correlations in the perpendicular direction. As the percolation threshold is approached from below, clusters become anisotropic in shape with a parallel correlation length, $\xi_{\parallel}$, diverging as $(p_+ - p_{+_c})^{-\nu_{\parallel}}$, and a perpendicular correlation length, $\xi_{\perp}$, diverging as $(p_+ - p_{+_c})^{-\nu_{\perp}}$, with $\nu_{\parallel} \neq \nu_{\perp}$. Accounting for this anisotropy in the Ginzburg criterion, we find that d_c, the upper critical dimension for directed percolation, equals five, compared to $d_c = 6$ for isotropic percolation.

We have also shown that $\xi_{\perp}$ does not behave as a true length under rescaling, but rather as a length *times* an angle (Klein and Kinzel 1981, Klein 1982b). Specifically,

$\xi_{\|}$ and $\xi_{\perp}$ are related by $\xi_{\perp} = \xi_{\|}\theta$ where θ is the opening angle of directed percolation clusters. This relation between the correlation lengths has important implications for the construction of a renormalization group treatment of this problem.

In addition, we have constructed a theory for directed percolation which is in the spirit of the Flory treatment of excluded-volume effects for linear polymers (Redner and Coniglio 1982, Lubensky and Vannimenus 1982). It predicts the correct upper critical dimension of $d_c = 5$, and gives the analytic dimension-dependent expressions of $\tilde{\nu}_{\|} = (d+9)/4(d+2)$ and $\tilde{\nu}_{\perp} = 7/4(d+2)$, where $\tilde{\nu}_{\|}$ and $\tilde{\nu}_{\perp}$ describe the divergence of the correlation lengths on N, the average number of bonds in a cluster. The Flory values are remarkably accurate in two and three dimensions where numerical data exists, and makes predictions for higher dimensions which await numerical tests.

We have developed a cell PSRG approach which incorporates the directionality effects of the diodes. For the square lattice, we obtain the phase diagram which reveals a wide variety of geometrical behavior. There are two second-order surfaces emanating upward from a central line to form a wedge-shaped region. Within this volume lies the positive diode phase where percolation in only one direction can occur. An identical structure occurs below the reflection symmetry plane so that the diagram is divided into four regions: the positive diode, negative diode, resistor, and insulating phases. Here resistor and insulator refer to isotropic percolation and no percolation, respectively. On the simple cubic lattice, a qualitatively similar phase diagram is obtained. A number of novel percolation transitions are predicted by our approach:

(i) Directed percolation. Directed percolation corresponds to the $p_+ - q$ (or $p_- - q$) axis of the phase diagram. As already mentioned, two independent diverging lengths, $\xi_{\|}$ and $\xi_{\perp}$, are required to characterize cluster shapes. Above the threshold, percolation is confined within a narrow cone whose opening angle ϕ scales as $\xi_{\perp}/\xi_{\|}$. Because of the anisotropy, novel PSRG approaches are required. We have developed such a PSRG in order to study directed percolation quantitatively (Herrmann et al 1983, see also Zhang and Yang 1984). These new approaches are an important first step in treating anisotropic critical phenomena through the renormalization group.

(ii) "Reverse" percolation. This novel transition occurs as a lattice completely occupied by one species of diode is gradually diluted by resistors. With no resistors present, one quadrant of the lattice is "wetted" by a fluid source at the origin. As the concentration of resistors increases, the angle of this percolating region increases from 90^o to 180^o just

below the transition, and approximately half the lattice is wetted. Above the transition, an infinite "backflow" path forms and the entire lattice is wetted.

(iii) More general percolation transitions. Our network model also displays novel percolation transitions in which two parameters, one related to the bond concentration, and the other related to the average bond orientation, can drive a percolation transition. One transition retains the character of the usual isotropic percolation threshold. This transition occurs when the total concentration of bonds increases to a critical value, while the average orientation of any diodes present remains random. One important example that falls in this class is "random Manhattan"—a lattice completely occupied by randomly oriented diodes. This network is obtained if all the one-way signs in midtown Manhattan were randomized at every intersection. By the use of the PSRG and an exact analysis of the pair-connectedness function, we have argued that random Manhattan is *isomorphic* to pure bond percolation at its threshold. This result also holds for the intermediate situation of a network containing resistors as well as randomly-oriented diodes.

A second independent transition occurs as the diode "polarization," $h = p_+ - p_-$, is varied. As $h \to 0$, the critical point is approached from one of the diode phases, and length of backflow paths opposite to the diode polarization diverges.

Finally, at the isotropic percolation threshold we have proved the equality between a number of exponents which holds for all lattices. This result reflects a simple geometrical relationship between the number of isotropic and directed "cutting" bonds in the network. This insight is a first step in constructing an intuitive picture of the geometrical structure of random resistor-diode networks.

Conduction Properties of Resistor-Diode Networks

We have investigated the conduction properties of electrical networks in which some of the circuit elements may have an asymmetric current-voltage response. Such a situation should describe certain features of strong-field hopping conductivity in doped semiconductors. As an intial step in understanding these complicated systems, we have introduced and studied simple idealized networks whose properties are dominated by directionality effects. An example is a network containing "ohmic" diodes of a fixed spatial orientation. Such a circuit element behaves as an ideal resistor under a forward-biased voltage, and is non-conducting under back-biased conditions. Experimentally, this has been achieved in an approximate way by a series combination of a real diode and resistor (Redner and Brooks 1982). We have also extended our study to more general situations in which the conductivities of the asymmetric elements in the forward and reverse directions, σ_+ and σ_- respectively, may be either infinite, finite, or zero.

(a) Mean-field theory

The mean-field limit for the directed conductivity exponent may be found by applying the de Gennes-Skal-Shklovskii links and nodes model, which provides an idealized geometric picture for the percolating cluster. For pure percolation, the infinite cluster can be represented by a regular array of nodes of mean separation ξ, joined by links whose conductances vanish as $(p-p_c)^\zeta$, when $p \to p_c$ from above. The conductance of a d-dimensional network of linear dimension L can then be obtained by superposing the $(L/\xi)^{d-1}$ chains, each containing L/ξ links. This leads to a network conductance that varies as $L^{d-2}(p-p_c)^t$, with $t = \zeta + (d-2)\nu$.

For directed percolation, the node lattice becomes anisotropic, with spacings of $\xi_\parallel$ and $\xi_\perp$ respectively, parallel and perpendicular to the anisotropy axes of the system. This modification leads to a directed conductivity exponent $t_+ = \zeta_+ + (d-1)\nu_\perp - \nu_\parallel$. Employing the values $\zeta_+ = 1$, and $\nu_\parallel = 1$, $\nu_\perp = 1/2$ valid for $d \geq d_c = 5$, we obtain $t_+ = 2$, compared to $t = 3$ for the mean-field limit of the random resistor network, valid at six dimensions and above. The exponent inequality $t_+ < t$ indicates that the directed conductivity should have a much sharper variation with bond concentration near the percolation threshold. This is intuitively plausible since the long tortuous paths that cause a very small conductivity in the random resistor network, cannot occur in directed percolation. This observation leads to a number of experimental ramifications, most notably the phenomenon of negative resistance in strong-field hopping conductivity (Böttger and Bryksin 1982).

(b) Analog experiments

As a more direct approach to studying directed conductivity, we have performed analog experiments on the directed network (Redner and Brooks 1982, see also Arora et al 1983). At first sight, it does not appear possible to extract information about the directed conductivity exponent because of the large jumps in the conductivity as a function of the number of bonds cut. These jumps are strongly correlated with the size of the underlying directed backbone, however, and we can use this information to obtain an estimate for t_+. The source of these jumps are the topological constraints of directed pecolation, where the removal of a single bond disconnects a large portion of the network. In contrast, if the backbone in this figure consisted only of resistors, removing the indicated bond in the figure, would entail the removal of only four additional bonds to obtain the new backbone.

The strong correlation between the backbone size and the conductivity shows that the two quantities are nearly proportional. The directed backbone exponent β' is a purely geometrical quantity for which it is known that $\beta' = 2\beta$ exactly. When coupled with the numerical estimate $\beta = 0.28$, we estimate $t_+ \approx 0.6$.

(c) Computer simulations

In a network containing diodes, there is the possibility that some of the diodes are back-biased, and hence do not contribute to the conductivity even though they may be part of the geometrical backbone of the cluster. Consequently, new methods need to be developed to first identify the subset of bonds which are forward-biased, and then determine the conductivity.

We have developed a numerical iteration and relaxation method which accomplishes this task (Redner and Mueller 1982). To explain the method, note that an obvious extension of relaxation is to allow the state of the network to change at each iteration step. Back-biased diodes should turn off, while previously turned-off diodes should turn on again if the voltage across them becomes forward biased. Thus the state of the network should "float" during the calculation. This procedure leads to prohibitively slow convergence in many cases, and an apparent limit cycle behavior in some pathological cases. The reason for this oscillatory behavior stems from the potential for negative feedback between the states of certain nearly balanced diodes.

To overcome this problem, we developed a more gradual relaxation method in which we effectively smooth out the break in the I-V response of each diode at $V = 0$ by replacing the response curve with a piecewise continuous function. With this method, oscillations are greatly reduced, and much more rapid convergence to the correct conductivity is obtained.

To estimate the conductivity exponent, we have developed a novel *anisotropic* finite-size scaling method. Due to the anisotropy of directed percolation, the linear dimensions of the system parallel and perpendicular to the anisotropy, $L_{\parallel}$ and $L_{\perp}$ respectively, must scale up according to $L_{\parallel}^{1/\nu_{\parallel}} \simeq L_{\perp}^{1/\nu_{\perp}}$. Very approximately, if the width of the system doubles, the length must triple. Accordingly, we study a sequence of lattices beginning at a small size such as $L_{\perp} \times L_{\parallel} = 1 \times 2$, 1×1, or 2×1, and scaling up to 32×478, 48×453, or 48×152 respectively. Only under these conditions will the conductivity scale as $L_{\parallel}^{t_+/\nu_{\parallel}}$. Based on our Monte Carlo data obtained at the percolation threshold, we estimate a directed conductivity exponent of $t_+ = 0.60 \pm 0.10$, in good agreement with the analog experiment.

(d) Position-space renormalization group (PSRG)

We have developed a cell PSRG approach for directed conductivity by rescaling both the bond occupation probabilities and the bond conductivities. One advantage of the PSRG treatment is that it can be extended straightforwardly to treat more general circuit elements such as superconducting bonds and also more complicated geometries such as randomly-oriented diodes. At present, we have considered an "oriented" resistor-diode network, which contains resistors, vacancies, and one species of ohmic diodes (Redner 1982c).

Such a network has three phases, depending on whether the forward or reverse conductances, G_+ and G_- respectively, are zero or non-zero. In the diode phase, G_+ is non-zero, and it vanishes according to the exponent t_+ as the boundary with the non-conducting phase is approached. On the other hand, in the resistor phase, both G_+ and G_- are non-zero, and only G_- vanishes as the boundary with the diode phase is approached. The vanishing of G_- may be written as $(\delta p)^{t_-}$, where t_- is a "reverse" conductivity exponent, and δp *is the distance from the diode phase boundary. Finally, at the isotropic percolation* point, both G_+ and G_- vanish, and we have a tricritical point with two independent conductivity exponents. One is simply the isotropic conductivity exponent which describes how G_+ and G_- simultaneously vanish as the transition is approached from the resistor phase. There is a second exponent which describes how G_+ only vanishes as the transition is approached from the diode phase. From a $b = 2$ rescaling we have calculated these exponents, and in particular, our result for t_+ is in good agreement with our numerical approaches discussed above.

In addition, a similar PSRG calculation can be used to find the conductivity divergence of an ohmic diode-superconducting diode mixture. The PSRG requires that the parameter space be enlarged to describe the network self-consistently. From this approach we predict a wide variety of network responses characterized by the interplay between ohmic conductivity and superconductivity, and also directionality effects.

C. Transport in Low-Density Materials

How are the laws of physics for this new class of low-density materials discussed above? For example, how are the conventional laws of diffusion and flow modified in a randomly porous medium? This question has been of the highest practical importance for some time, yet it is only in very recent months that substantial progress has occurred.

The main idea is the recognition of two time scales, with the borderline depending on the characteristic linear dimensions of the inhomogeneities in the randomly porous material. For a conventional Euclidean system, there is only a single time scale. For any value of the time t, Fick's law applies: the rms displacement of a particle $\xi_p = \langle r^2 \rangle^{1/2}$ varies as $t^{1/2}$. For a fractal structure, there are *two* time domains. If we wait long enough, Fick's law will apply and $\xi_p \sim t^{1/2}$. However for short times the range of the diffusing particles, ξ_p, is shorter than the characteristic length scale ξ_f characterizing the fractal. In this time domain, Fick's law breaks down, and the rms displacement varies with time according to a completely different power law, $\xi_p \sim t^{2/d_w}$.

The parameter d_w is called the fractal dimension of the random walk that the diffusing particle undergoes in a porous material. The special case $d_w = 2$ corresponds to diffusion in a non-porous medium. One remarkable discovery is that while d_w *always* has the value of 2 for a non-porous medium [regardless of the dimension of the space, or other details], for a porous medium d_w depends very strongly on d.

It is often customary to discard data taken at short times, since these data do not obey Fick's law. Now we understand that these data follow a quite different behavior which yields valuable information about the nature of the porous medium. It is therefore important to re-analyze a wealth of data in light of this development, and to examine carefully all the implications of this new law of diffusion and flow. In particular, we can calculate the fraction of material "wetted" by the diffusion process, and we find here that there is again a remarkable new law emerging for randomly porous media. Moreover, we can elucidate the behavior of a randomly porous system in a velocity field. The flow equations are also modified substantially by the fractal structure of porous media, for all but the longest time scales.

In discussing transport in random media, another basic question is, "what happens for *high* flow rates?" In this case, it appears that transport processes become anisotropic in character. To be specific, consider placing a diffusing particle within a porous medium in which fluid is flowing rapidly. In this case, the particle will drift along the field, while performing Brownian motion transverse to it. We propose to explore the quantitative laws that describe this anisotropic transport process. In particular, for a homogeneous medium, the displacement along the flow will vary linearly with time, while the transverse diffusion will be governed by Fick's law. However, just as for isotropic problems, the randomness modifies these scaling laws in a fundamental way.

In addition to the aforementioned anisotropy, strong flow fields also suggest the existence of ***non-linear*** effects. It is only for weak flows that transport can be described by linear response theories. For strong flows, collective particle motions become important, and flow rates depend non-linearly on biasing fields. In particular, negative differential resistance has been observed in doped semiconductors for an appropriate range of doping and field strengths. As mentioned in Sec. B above, these interesting effects can be described simply in terms of a discrete random network model in which transport is mediated by one-way bonds or diodes.

In order to test the theoretical ideas of diffusion on randomly porous structures, it is necessary to be able to characterize the morphology of the medium accurately. As mentioned earlier, the characteristic size scale ξ_f determines an important crossover effect. For $\xi_p > \xi_f$, Fick's law holds, while for short times, Fick's law breaks down. One medium in which to observe the breakdown of Fick's law and measure transport properties in the short time regime is a polymer gel. By varying the composition of the monomeric units of various functionalities that comprise the gel, it is possible to vary the characteristic mesh size of the gel over a wide range. This mesh size plays the role of ξ_f in the diffusion measurements. Furthermore, Professor R. Bansil here can measure the mesh size quite accurately by using macromolecules of known molecular weight and size and studying the permeability of these macromolecules in the gel. Other techniques to measure mesh size such as small angle light scattering will also be considered. Thus by performing tracer diffusion experiments on gels with varying mesh sizes, we propose to test the theoretical ideas of diffusion in random media. It is also possible to study the diffusion of polystyrene microspheres of known size in a medium which is undergoing gelation so that the characteristic length ξ_f is changing with time. In this way one can study the crossover from diffusion in a *homogeneous* solution to diffusion in a fractal.

We have preliminary results that contribute to our confidence that meaningful progress can be achieved. Many of these results revolve around the discovery of Alexander and Orbach (1983) that the parameter d_w introduced above to characterize diffusion in a randomly porous media is directly proportional to the fractal dimension d_f of the material,

$$d_w = \frac{3}{4} d_f. \tag{3}$$

We have concentrated our initial efforts on two main questions:

1. Is the Alexander-Orbach conjecture exact or only approximate? In an effort to do this, we have introduced a function $G(p, N)$, which is the number of growth sites of a percolation cluster after N steps of the walk. The cluster is imagined to be generated as the walker moves about, by flipping a coin each time the walker considers the possibility of visiting a new site of the lattice; if the coin comes up heads then the walker moves to that site while if the coin comes up tails then the walker treats that site as blocked "forever." The coin is weighted to come up heads with probability p, and growth sites are those unblocked sites that are neighbors of visited sites. It appears that the function $G(p, N)$ plays the role of the "order parameter" in the problem of diffusion in porous media (Coniglio et al, to be published). In particular, $G(p, N)$ approaches zero as $p \to p_c$ from above. Exactly at p_c $G(p_c, N)$ may be written as a sum of independent random variables for the case of the Cayley tree, from which the Alexander-Orbach relation follows rigorously (Leyvraz and Stanley 1983). We propose a thorough and careful investigation of the function $G(p, N)$ and the modifications in the Cayley tree argument that would be needed to justify the Alexander-Orbach conjecture for general d-dimensional Euclidean lattices. It is of the greatest importance to learn if the Alexander-Orbach conjecture is exact or only approximate. Arguments based on epsilon expansions suggesting that it fails just below six dimensions have been criticized (Coniglio 1983), and increasingly the numerical evidence suggests that it may hold (to at least 2-3%) for *all d*. Of course, it is possible that it is "like the Flory theory" and holds for some values of d and not others, so we propose a careful Monte Carlo study of d-dimensional percolation using extremely large system sizes and extremely long random walks. Such a program has been initiated independently by the Toulouse group but we are certain that we can obtain more accurate numbers since our own computer resources are quite immense (for example, we have succeeded in simulating percolation clusters of up to 17 billion sites!).

2. To what class of fractals does the Alexander-Orbach conjecture apply? The above remarks concern the percolation fractal, a model of randomly porous media just at the threshold of conduction. There are many other low-density materials of great current interest. Once such example is the model of colloids called diffusion-limited aggregation, proposed by Witten and Sander (1981). In this model, the fractal is created in an irreversible fashion by allowing particles to diffuse in from a large distance and to stick to the growing aggregate whenever they touch it. The resulting Witten-Sander aggregates are large, wispy and highly ramified, with a fractal dimension that depends strongly on d.

Recently we have initiated an extensive study of diffusion on Witten-Sander fractals (Meakin and Stanley 1983). Our preliminary results for $d = 2, 3$ suggest that (4) holds to an accuracy of at least 10%. We are currently increasing the accuracy, as well as extending these studies to all dimensions. In order to increase the accuracy, we have begun using a clever idea of Havlin (1983, unpublished). Normally, computer simulations are characterized by two statistical averages: first one averages over many fractals, and second one averages over many walks on each fractal. However the second average can be eliminated! Specifically, we can now enumerate *exactly* the probabilistic features of the random walk on the fractal by calculating analytically the probability that the end point of the walk is at position r after N_w steps. We find a dramatic increase in the accuracy using this method, and propose to apply it to a wide range of low-density structures in addition to Witten-Sander aggregates.

We have also begun to study the question of whether the Alexander-Orbach conjecture applies to a completely different class of phenomena, diffusive annihilation. Here one studies a system of particles that are free to diffuse at random in a continuum, but if two particles touch then they annihilate. One monitors the surviving density of particles as a function of time, and finds that the decrease is not exponential but rather via a long-time power law whose exponent is d_f/d_w (Meakin and Stanley 1984, Kang and Redner 1984). Preliminary simulations in $d = 2, 3$ show that (3) is obeyed quite accurately. These results show that (3) is far more general than first imagined.

The remarkable generality of (3) thus motivates one to wonder if there are any systems for which it does not apply. There do seem to be such systems. These include non-random geometric fractals such as the Sierpinski gasket and "Havlin" carpet, as well as a most interesting and relevant fractal: the percolation backbone. We have been able to show rigorously (Stanley and Coniglio 1984) that

$$\bar{d}_w - \bar{d}_f = d_w - d_f, \tag{4}$$

where the bars denote the backbone. Moreover, we have found that all numerical evidence is consistent with the possibility that for the backbone (3) is replaced by a relation involving the exponent characterizing the resistance to flow of fluid at the critical point.

Thus we are left with a most puzzling physics problem: (3) applies to some fractal materials and not to others. How can one predict in advance of actual calculations whether it will apply? I.e., what are the features of a fractal that cause it to obey (3)? One possible answer to this question has been suggested recently (Leyvraz and Stanley 1983), namely that homogeneous fractals obey (3) while non-homogeneous fractals do not. By homogeneous we mean that there are no bottlenecks that hinder the diffusion. More precisely, for a homogeneous fractal the number of elements of a set of sites that totally surrounds a fractal scales as $d_f - 1$. In contrast, nonhomogeneous fractals such as the Sierpinski gasket and Havlin carpet have boundary sets with exceptionally dense boundaries (hence the term nonhomogeneous). We propose to consider other possible categorizations in an effort to find the best one.

In summary, the Alexander-Orbach discovery, (3), has far-reaching implications for our understanding of transport in randomly porous materials [among which is that dynamic critical exponents are related to static exponents–e.g., the electrical conductivity exponent t is given by $t/\nu = d-2+\frac{1}{2}d_f$. Accordingly, we propose to bring to bear on this problem all methods of statistical mechanics, such as renormalization group, Monte Carlo simulation, and exact enumeration. Very recently we have begun to consider transport by mechanisms other than diffusion. One such is the transport occurring on a superconducting network, where we have succeeded in deriving a relation between the transport exponent s/ν and the fractal dimension, analogous to that quoted above for the exponent t (Coniglio and Stanley 1984).

D. Non-Equilibrium Properties of Low-Density Materials

Properties of materials depend strongly on the process of formation as well as chemical composition. In addition, several classes of useful materials are not in the equilibrium state, but are in a glass or metastable state. For these reasons we have begun to study non-equilibrium properties of materials.

There exists a substantial body of research demonstrating that percolation concepts are essential for understanding phenomena such as nucleation (Klein and Unger 1983), spinodal decomposition (Binder 1983), and glasses (Grest and Cohen 1983). In particular, the problems of continuum percolation (where there is no underlying lattice structure) and correlated percolation (where the percolating elements interact) play an essential role. In this section we discuss our previous and proposed work in these areas and indicate their relevance to non-equilibrium processes such as nucleation as well as to other practical problems such as fuel cells.

1. Continuum percolation

Some of the areas on which percolation theory has a substantial impact include design of liquid electrolyte fuel cells, secondary and tertiary oil recovery, properties of polymeric materials and effect of impurities on metallic alloys. In a substantial fraction of these areas the problem of percolation in a continuum, as opposed to percolation on lattice structures, contains the essential physics. For this reason we have an ongoing program involving computer simulation, renormalization group and rigorous results in which we are studying continuum percolation and its application to various practical problems. In the following we describe several of these projects in some detail, outlining the progress that has been made. In the projects below various types of percolation will be described (e.g., correlated and uncorrelated continuum percolation) and several methods will be outlined. The one unifying factor is that in all of the projects the objects whose connectivity properties are being studied are free to move throughout d dimensional space and are not constrained to be on a fixed regular lattice.

(a) Computer simulations and Monte Carlo renormalization group

An essential problem in percolation is "what effect does the lattice have on the underlying connectivity properties?" That is, "what aspects of the vast amount of information known about lattice systems can be applied to the continuum?" One aspect of this question is the problem of whether lattice and continuum percolation have the same critical exponents. To investigate this problem we have performed Monte Carlo simulations (Gawlinski and Stanley 1981) and Monte Carlo renormalization group studies (Gawlinski and Redner 1983) of non-interacting squares and squares that have an additional hard core repulsion. Our results indicate that for such systems critical exponents remain unchanged from the values obtained in lattice models.

Realistic systems, however, have attractive forces between molecules. There is evidence (see next section) that near a phase transition, critical percolation behavior is strongly modified. We are currently investigating this effect in continuum percolation. Other questions, which we will investigate with Monte Carlo techniques in continuum systems, are

[1] The structure of the percolating network.

[2] The size distribution of clusters.

[3] The relationship between cluster properties and current flow in disordered media and also fluid flow in randomly porous media.

The answers to the above questions will provide a basis for investigating structural and electronic properties of porous low-density materials.

(b) Potts model formulation

Numerical methods have so far provided the only insights into continuum percolation. Theoretical investigations are hampered by the complexity of continuum systems compared to lattice models. We began a promising approach based on a major result of Kastaleyn and Fortuin (1969) who demonstrated that the quantities of interest in lattice percolation, a problem that considers connectivity can be obtained by solving for the thermal properties of a spin model, the s-state Potts model. The advantage of this breakthrough is that methods such as renormalization group developed for lattice thermal problems could now be applied to lattice percolation problems.

We have generalized this mapping from thermal problems to connectivity problems to include continuum models (Klein 1982a). This mapping has made techniques available for continuum percolation which have been developed to solve continuum fluid problems.

Several projects have exploited these insights. These include:

[1] We have used this "Potts model" formalism to derive hierarchies of integral equations for the connectivity functions (Klein and Stell 19xx). These functions play the role in percolation that the distribution function play in the theory of fluids. With these hierarchies we can derive, again in analogy to fluid theory, approximate integral equations that will describe the connectivity properties in continuum percolation over a wide range of densities and correlation parameters.

[2] We have developed a method of solving linear integral equations with renormalization group techniques (Klein 1983). This method is being applied to some of the heirarchies and approximate integral equations mentioned above.

[3] The model described above is applicable to the problem of percolation of spheres. This is necessary for understanding problems such as electron flow in fuel cell matricies. For the problem of gaseous diffusion, electrolyte transport in fuel cells or oil recovery the problem one needs to understand is percolation of the voids, the spaces between the spheres. We are studying ways to adapt our model to this important problem.

2. Correlated percolation

Although random percolation, either on a lattice or in the continuum, has important applications most of the systems occuring in nature have important correlations. In certain cases, such as systems near phase transitions, the interaction or correlation is such an essential part of the physics that random models are irrelevant or misleading. During the past few years we have developed many correlated percolation models that have been useful in describing phenomena such as critical behavior in Ising and Potts models, nucleation in deeply quenched metastable states and anomalous behavior of water. Below we describe our progress in developing models for the above phenomena, various methods we have developed to solve these models and outline our present and future projects.

(a) Correlated percolation near the critical point. Although short-ranged correlations have very little effect on connectivity properties at the percolation threshold, the situation is markedly different at thermal phase transitions (Klein et al 1978, Coniglio and Klein 1980, Coniglio and Lubensky 1980, Tuthill and Klein 1982,1983). At thermal phase transitions such as critical points the correlation length becomes infinite and *percolation* critical exponents become modified by the *thermal* fluctuations. This phenomenon has been extensively studied in ferromagnetic Ising models (Klein et al 1978, Coniglio and Klein 1980, Coniglio and Lubensky 1980, Jan et al 1982, Weinrib and Halperin 1983, Benzoni and Cardy 1983). Results on the Bethe lattice (Coniglio et al 1977,1979,1982), series analysis (Sykes and Gaunt 1976), and Monte Carlo simulations (Stauffer 1981, Heermann and Stauffer 1981, Ottavi 1981, Roussenq et al 1982, Kertész et al 1983) are available. In addition, investigations were also made of antiferromagnetic Ising models (Amitrano et al 1983) and the Potts model (Coniglio and Peruggi 1982). From these extensive investigations the picture that emerges is that in the neighborhood of critical points percolation transitions can not only be modified but that a mathematical mapping exists between percolation and and various thermal phase transitions. This mapping allows one to identify thermal fluctuations with percolation clusters so that concepts such as the fractal dimension (Mandelbrot 1977) can be applied to thermal problems. It has also led to an improved definition of droplets in metastable states (see discussion below). Although a great deal has been understood over the past five years, several important questions need to be answered.

Nucleation

The standard or classical theory of nucleation assumes that the metastable phase (e.g., a supercooled gas) decays into the stable phase (e.g., liquid) due to the occurence of large enough droplets of the stable phase which occur in the metastable background. These droplets are assumed to be compact; that is, the fractal dimension of the droplet d_f is assumed to be equal to the dimension of space (i.e., $d_f = 3$ for experiments). Therefore the volume of the droplet is proportional to r^d and its surface to r^{d-1}. Computer simulations (Stauffer et al 1982) of nearest-neighbor Ising models have tested the classical theory far from the critical point and found data consistent with the theory. For Ising models with longer range interactions, however, computer simulations (Heermann et al 1982, Heermann and Klein 1982) and theory (Klein 1981, Klein and Unger 1983) have shown that the nucleating droplets are not compact but are ramified and the surface of the droplet is proportional to its volume. This is also true near the critical point (Heermann et al 1983).

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PUBLICATIONS UNDER THE PRESENT GRANT

[*Denotes articles with ARO support.]

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